OPTICAL STUDY OF LIGHT EXTRACTION IN CORRUGATED ORGANIC LIGHT EMITTING DIODES

By

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To my parents and my wife
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<td>AFM</td>
<td>Atomic Force Microscope</td>
</tr>
<tr>
<td>Alq3</td>
<td>Tris(8-hydroxy-quinolinato) aluminum</td>
</tr>
<tr>
<td>EL</td>
<td>Electroluminescence</td>
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<td>EML</td>
<td>Emitting Layer</td>
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<td>EQE</td>
<td>External Quantum Efficiency</td>
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<td>EBL</td>
<td>Electron Blocking Layer</td>
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<td>EIL</td>
<td>Electron Injection Layer</td>
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<td>ETL</td>
<td>Electron Transport Layer</td>
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<td>Hole Blocking Layer</td>
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<td>HIL</td>
<td>Hole Injection Layer</td>
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<tr>
<td>HTL</td>
<td>Hole Transport Layer</td>
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<tr>
<td>HOMO</td>
<td>Highest Occupied Molecular Orbital</td>
</tr>
<tr>
<td>IQE</td>
<td>Internal Quantum Efficiency</td>
</tr>
<tr>
<td>Ir(ppy)$_2$(acac)</td>
<td>Bis<a href="2,4-pentanedionato-O$_2$O$_4$">2-(2-pyridinyl-N)phenyl-C</a>iridium(III)</td>
</tr>
<tr>
<td>ITO</td>
<td>Indium Tin Oxide</td>
</tr>
<tr>
<td>LUMO</td>
<td>Lowest Unoccupied Molecular Orbital</td>
</tr>
<tr>
<td>OLED</td>
<td>Organic Light Emitting Diode</td>
</tr>
<tr>
<td>PEDOT:PSS</td>
<td>Poly(3,4-ethylenedioxythiophene):polystyrenesulfonate</td>
</tr>
<tr>
<td>PDMS</td>
<td>Polydimethylsiloxane</td>
</tr>
<tr>
<td>TADF</td>
<td>Thermally Activated Delayed Fluorescence</td>
</tr>
<tr>
<td>TAPC</td>
<td>di-[4-(N,N-ditoly-amine)-phenyl]cyclohexane</td>
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OPTICAL STUDY OF LIGHT EXTRACTION IN CORRUGATED ORGANIC LIGHT EMITTING DIODES

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The key factor limiting the external quantum efficiency of OLED devices is the outcoupling efficiency. For a planar OLED device, the outcoupling efficiency is only 20% to 30\%^{1,2}. A large portion of light is trapped in substrate mode, waveguided mode and SPP mode\^{2,4}. Researchers have studied many light extraction techniques to recover light from these loss channels, including corrugation\^{5,6}, macro-lens\^{7}, micro lens arrays\^{3}, roughened substrate\^{8,9}, sub-anode gridding\^{10}, scattering layer\^{11} and nano-composite substrate\^{12}.

This thesis stands from an optics point of view to understand light extraction with corrugation, one of the most effective way to extract waveguided mode and SPP mode\^{11}. For a corrugated OLED, a photonic crystal pattern is first replicated by imprinting to form a corrugated substrate, then OLED stack is sputtered/evaporated on the corrugated substrate. Due to Bragg diffraction of the corrugation, the trapped waveguided mode and SPP mode are extracted from the device. Corrugated OLEDs
show a 32% enhancement in current efficiency and 22% enhancement in EQE compared to the planar device.

Two kinds of corrugation patterns, single photonic crystal and poly photonic crystal are compared in this research. The single crystal OLEDs have distinguished diffraction features from extracted waveguided and SPP modes, which helps to understand the interaction between corrugation and OLED device structure. The poly crystal corrugation overcomes the angular dependent EL spectrum and strong glare issues which limit the application of single crystal corrugation.

The corrugated OLED devices are studied with angle-revolved electroluminescence spectroscopy (ARES). ARES precisely maps out the device air mode, traces back the origin of extracted light to waveguided and SPP modes, and allows quantification of the light extraction amount. In-depth analysis of the effect of corrugation depth and index contrast reveals the extraction efficiency can be further improved by maximizing corrugation depth while maintaining electrical stability, and increasing the index contrast at corrugated interfaces.
CHAPTER 1
FUNDAMENTALS OF ORGANIC LIGHT EMITTING DIODES

Background

Since Ching W Tang and Steven Van Slyke published the milestone paper on OLED in 1987, it has been studied intensely from various aspects, including electroluminescence mechanism, charge transport properties, lifetime, optical structures, in-expensive processing techniques etc. OLED emitting material has seen the breakthrough of fluorescence, phosphorescence and thermally activated delayed fluorescence. The external quantum efficiency of a planar device has been improved from 1% to over 30%, luminous efficiency has been improved from 1.5 lm/W to over 100 lm/W, device lifetime has been improved from a couple of hours to over a million hours. OLED techniques can be seen in televisions, cell phones, virtual reality wearable devices and lighting applications.

The working principle of a simple OLED device can be described as such: a luminescent small molecule (emitter) is doped in another organic matrix (host) to form the emitting layer (EML). The EML is sandwiched by the electron transporting layer (ETL) and hole transporting layer (HTL). ETL is connected to a negative bias by a cathode, and HTL is connected to a positive bias by an anode (Figure 1-1). Electrons are injected from cathode, transported by the ETL to the EML; holes are injected from anode, transported by the HTL to the EML. Electrons and holes recombine in the EML, excite the emitter from ground state to excited state, and form excitons. Excitons relax to the ground state, and emit photons through radiative decay.

The simple OLED structure mentioned before usually cannot achieve full utilization of charges. Therefore, additional layers are used in more complicated OLED structures.
(Figure 1-1)\textsuperscript{16,29-31}. A hole injection layer (HIL) can be inserted between anode and HTL to inject holes, and an electron injection layer (EIL) can be inserted between cathode and ETL to inject electrons. An electron blocking layer (EBL) can be inserted between EML and HTL to confine electrons in the EML, and a hole blocking layer (HBL) can be inserted between EML and ETL to confine holes in the EML. EBL is usually a HTL layer with high LUMO level to prevent electrons from leaving the EML. HBL is usually an ETL layer with low HOMO level to prevent holes from leaving the EML. These function layers are often thinner than 10 nm, with EIL and HIL usually thinner than 2 nm.

Although it seems an OLED device is a complicated structure which consists of many layers, each with distinct functions. From the optical perspective, an OLED is simply made of two components: the electroluminescent material (emitter), and the rest of the layers supporting the emitter (matrix). We now discuss about how these two components determine the efficiency of an OLED.

**Emitter**

The thickness of a typical OLED device is around 300~400 nm. The thickness of the EML is 20 to 30 nm. The doping ratio of the emitter in the EML is between 5 to 10\%\textsuperscript{32}. This means the emitter only makes up for less than 1\% of the OLED device. However, the emitter is the ultimate component that determines the color and maximal efficiency of the OLED device.

Emitters can be categorized by their emission mechanisms. When electrons and holes recombine, the ground state ($S_0$) of the emitter is excited to form two kinds of excitons: singlets and triplets. In most of the cases, only the first excited states ($S_1$ or $T_1$) can be reached\textsuperscript{33}. According to spin statistics, singlets and triplets are generated in a ratio of 1:3\textsuperscript{34}. The singlet states have a total spin $S = 0$, and the triplet state has a total
spin $S = 1$. Due to the spin state, excited singlets ($S_1$) can radiatively relax to $S_0$, while excited triplets ($T_1$) normally cannot radiatively relax to $S_0$. The utilization of excitons is called branching ratio ($\gamma$).

In 1987, Ching Tang and Van Slyke demonstrated the first fluorescent OLED\textsuperscript{13}. In this device, the singlets can go through fast radiative decay and give out fluorescence. While the radiative decay of triplets is very slow because $T_1$ and ground state have different spins. Most of the triplets end up generating heat. In this scenario, only singlets are harvested to give out light, so the branching ratio of a fluorescent OLED is only 25%.

In 1998, Marc Baldo demonstrated a phosphorescent OLED that can harvest both singlets and triplets\textsuperscript{35}. In a phosphorescent emitter, a heavy metal induces spin-orbital coupling which causes the singlet and triplet states to mix. Due to the strong spin-orbit coupling, singlets can go through very fast intersystem crossing to $T_1$ state, and triplets can go through radiative decay to the ground state and give out phosphorescence. In this scenario, all the excitons are harvested, so the branching ratio is 100%.

Despite the high utilization of excitons in a phosphorescent OLED, there are still some challenges. First, the efficient phosphorescent emitters are limited to iridium and platinum complexes\textsuperscript{36-38}, which are fairly expensive. Second, the long lifetime of triplet makes it susceptible to several quenching mechanisms\textsuperscript{39}. This includes bimolecular quenching processes (triplet-triplet annihilation\textsuperscript{40}, singlet-triplet annihilation\textsuperscript{41}, polaron-triplet-quenching\textsuperscript{42} etc.). These quenching mechanisms can cause efficiency roll-off at high current density. In addition, triplets are easily quenched by host or adjacent charge transporting layers without triplet confinement\textsuperscript{43}, putting limitation in host and charge
transport material selection. In order to prevent triplet quenching, the triplet energy of host and charge transporting layers must be higher than that of the emitter, called triplet confinement\textsuperscript{43-44}. Without triplet confinement, the triplets generated in emitters will be quenched and will reduce the device efficiency. In blue phosphorescent OLED, because the triplet energy of a blue emitter is around 3 eV, which means HTL, ELT and host must have a higher triplet energy than the emitter. So far, making blue phosphorescent OLEDs with long lifetime and high efficiency remains a challenge\textsuperscript{23}.

One way to solve these problems is to use fluorescent OLED that can harvest triplets. In 2012, Chihaya Adachi demonstrated a triplet harvesting mechanism, called thermally activated delayed fluorescence (TADF)\textsuperscript{15}. In this process, triplets can go through reverse intersystem crossing (RISC) to the singlet state, and radiatively decay to the ground state and create delayed fluorescence. Since both singlets and triplets are harvested, a maximum of 100% branching ratio can be realized. The key to TADF is a fast RISC. Since it's a thermally activated process, the rate of RISC is dependent on temperature and the barrier height, which is the energy splitting between singlet and triplet energy.

Even with the matching spin states, an exciton can relax to the ground state through radiative or non-radiative decay path\textsuperscript{45-46}. The ratio of radiative decay over all the decay paths is called quantum yield ($\eta_{QY}$). Quantum yield can also be affected by the selection of host materials. Normally a host material has higher singlet, triplet energy than the emitter to avoid energy backflow. Most of the optimized OLED devices have a high quantum yield over 90%\textsuperscript{46}. 
Another way the emitter can affect the device efficiency is through emitter orientation\textsuperscript{46-50}. Consider the emitters as oscillating dipoles. When the dipole is lying horizontally, most of the light is emitted normal to the OLED thin film plane, which experiences less reflectance and total internal reflection; when the dipole is standing vertically, most of the light is emitted along the OLED thin film plane. Light travelling along the thin film is easily trapped in the waveguided mode, substrate mode and SPP modes. Any arbitrary emitter orientation can be decomposed into these two components. Therefore, it’s optically beneficial for the emitters to generate photons travelling in the normal direction.

The exciton energy level of the emitter also determines the color of the emitted photons. For fluorescent emitters, this means $E(S_1)$, and for phosphorescent emitters, this means $E(T_1)$. When exciton relaxes from $S_1$ or $T_1$ to the ground state through radiative decay, a photon with the energy equal to $E(S_1) - E(S_0)$ or $E(T_1) - E(S_0)$ is emitted. For OLEDs that emit visible light, the single or triplet energy of the emitter is usually between 1.9 eV and 2.7 eV.

**Matrix**

Other than the emitter, the rest of the OLED layers form a matrix. It is responsible for two major tasks, both electrical and optical.

From electrical point of view, the matrix is responsible for injecting and transporting charges to the emitter. The trapping rate of electrons and holes onto the emitter affects the utilization of charges, termed charge balance ($\eta_c$)\textsuperscript{49-50}. When one electron is trapped for each trapped hole, the charges are 100% balanced. When one kind of charges is excessive, $\eta_c$ decreases. Charge balance is achieved by carefully designing the OLED structure to optimize the injection and transport of both charges. The injection of
charges is affected by the energy level offset at an interface. For example, the difference between the work function of anode and the HOMO level of HTL\textsuperscript{51}, and the difference between the HOMO level of HTL and the HOMO level of EML\textsuperscript{52}. Smaller offset is beneficial for charge injection. The transport of charges is an intrinsic property of the layer, determined by its layer thickness and charge mobility ($\mu_e$ or $\mu_h$)\textsuperscript{53-54}. Doping with n-type or p-type dopants can alter the charge transport properties of a layer significantly\textsuperscript{55}.

From optical point of view, the matrix forms an optical cavity which affects the outcoupling of photons\textsuperscript{56-57}. The ratio of the photons that can emit into air over all the photons generated by the emitter is determined as outcoupling efficiency ($\eta_{\text{out}}$). A few factors can affect the outcoupling efficiency. Because organic layers ($n_{\text{org}} \sim 1.75$) have a higher refractive index than air ($n = 1$), they form an index waveguide which traps the photons\textsuperscript{2}. Also, most of the OLEDs uses a reflective metal as cathode (or as both cathode and anode), the emitted photons can couple with the charges of metal surface, and form surface plasmon polariton (SPP) modes\textsuperscript{21, 58}. Additionally, metal absorption\textsuperscript{59} and total internal reflection\textsuperscript{5, 60} can also reduce $\eta_{\text{out}}$. In a typical OLED device, the outcoupling efficiency is limited to 20~30%.

**EQE and Light Extraction**

The external quantum efficiency (EQE) of an OLED is defined as the number of photons emitted by the device for the number of charges injected. Considering only single unit OLED device, the maximum EQE achievable is 100%. The equation to calculate EQE is

$$\text{EQE} = \gamma \eta_{\text{QY}} \eta_c \eta_{\text{out}}$$

\text{(1-1)}
The products of the first three terms $\gamma \eta_{QY} \eta_c$ is usually defined as the internal quantum efficiency (IQE). With the design of high quantum yield phosphorescent or TADF emitters, in additional to the perfectly balanced charges, devices with close to 100% IQE have often reported. Because the $\eta_{out}$ for common planar OLED devices is only 20-30%, it has become the bottleneck of high efficiency OLED devices.

**Characterization of OLEDs**

OLED devices are characterized by their electrical and optical properties. Some of the common characterization methods are J-V-L measurement, EL spectrum measurement and EQE measurement.

In J-V-L (current – voltage- luminance) measurement, a voltage bias is applied to the anode and cathode of the OLED device through a Keithley 2400 SourceMeter. The SourceMeter also measures the driving current going through the OLED device. While driving the OLED device, a silicon photodiode (1.5 cm in diameter) is positioned close to the OLED pixel to convert light output into photocurrent. The photocurrent signal is picked up by a Keithley 6485 Picoammeter. The photocurrent is calibrated to the forward luminance through a LS-100 luminance meter. In this measurement, the OLED device is usually driven from -2 V to 8 V at a voltage step of 100 mV. The negative bias measures the reverse current of the device and provides information on the leakage current. To avoid fast degradation, the current compliance is set to 4 mA ($100 \text{ mA/cm}^2$).

EL spectrum measurement is done with an Ocean Optics HR4000 spectrometer. An optical fiber (200 $\mu$m diameter) is used to collect light from the OLED pixel, and the signal is processed by the spectrometer. The range of wavelength is from 195 nm to 1124 nm, which covers all the visible wavelengths for OLED study.
EQE measurement is done in a Labsphere Illumia integration sphere. The OLED device is positioned in the center of the integration sphere, and connected to a SourceMeter, a photodiode and a spectrometer. The interior of the integration sphere is designed so that light travelling in any directions will bounce multiple times and be collected by the photodiode. This process essentially ‘integrates’ all the light emitted by the device. The integration sphere is calibrated by a standard source, for which the spectral power distribution is known. We measure the J-V curve with the SourceMeter, and EL spectrum with the spectrometer. The photocurrent and EL spectrum are compared with the standard light source to calculate the total photon numbers at each driving voltage. The electron number at each voltage is calculated by the J-V curve. EQE can then be calculated by dividing the photon numbers with electron numbers.
Figure 1-1. The layer stack of a simple OLED device and a complex OLED device

Figure 1-2. Formation of singlet and triplet by electron-hole recombination
CHAPTER 2
OPTICAL SIMULATION OF OLED WITH SETFOS

Mode Distribution in OLEDs

A photon can be described by its energy (E) and wave-vector (k). For a free travelling photon in the air, \( E = h \nu = h c / \lambda, \) \( k_0 = 2\pi / \lambda. \) For a photon created by the emitter in an organic thin film, \( k \) equals \( n_{\text{org}} k_0. \) Using the organic thin film as the reference plane, \( k \) has an in-plane component \( k_x \) and an out-of-plane component \( k_z. \) \( k_x \) determines which optical mode a photon couples to. When \( k_x \) is smaller than \( k_0, \) the photon is emitted into air mode. In the air mode, \( k_x = k \sin \theta. \) \( \theta \) is the angle between the direction light travels and the normal direction of plane. \( k_x = 0 \) represents light that travels normal to the plane, and \( k_x = k_0 \) represents light that travels along the plane surface.

When \( k_x \) is greater than \( k_0, \) the photon is trapped in the OLED device. Consider a typical bottom emitting OLED device fabricated on a substrate (usually glass), and has a metal top electrode (usually Al or Ag). For \( k_0 < k_x < n_{\text{sub}} k_0, \) the photon is trapped in the substrate mode due to total internal reflection at the substrate/air interface. For \( n_{\text{sub}} k_0 < k_x < n_{\text{org}} k_0, \) the photon is trapped in the waveguided modes. The waveguide forms because the organic layers are sandwiched between a glass substrate, which has a lower refractive index than the organic layers, and a reflective metal electrode\(^2, 10, 61. \) For \( k_x > n_{\text{org}} k_0, \) the photon excites the charges on the surface of the metal electrode, and couples into SPP modes. Because \( k_x^2 + k_z^2 = k^2, \) \( k_z \) becomes imaginary for SPP modes, therefore SPP modes are evanescent.

Setfos is an optical simulation software developed by Fluxim\(^62. \) It calculates the light emitted in OLEDs using the dipole emission model, and calculates optical interference in thin-film layers. By inputting the layer thickness, and the optical constants including
refractive index and extinction coefficient of each layer, the optical cavity is established. Assume the emitter is in the center of the OLED organic layer stack, we can simulate the optical modes supported by the cavity.

Figure 2-1 shows a schematic drawing of an OLED device, and the major optical modes related to the structure. The Setfos simulation result of the same structure is also given. The simulated mode distribution plot maps out all the photon states by their energy (E) and in-plane wave-vector (kₓ). The z axis (color bar) represents the intensity of given photon state. The two dashed lines are drawn for guidance of eyes. The left dashed line represents the air line, which corresponds to the energy-momentum relation of a free travelling photon in the air (E = hν = ħck₀). From the E axis to the air line, the photons are emitted to air. The travelling direction of the photons changes from 0 degree (normal to the thin film plane) to 90 degrees (along the thin film plane).

The right dashed line represents the substrate cone, which corresponds to the energy-momentum relation of a free travelling photon in the substrate (E = hν' = ħck₀/n_sub). From the air line to the substrate line, the travelling direction of the photons in the substrate changes from critical angle θ_C = sin⁻¹(1/n_sub) to 90 degrees (along the substrate plane).

When kₓ increases further, the photons are trapped in the transverse magnetic (TM) waveguided modes, transverse electric (TE) waveguided modes or surface plasmon polariton (SPP) modes. Note that air mode and substrate mode are continuous, while waveguided and SPP modes are discrete along kₓ axis. The dispersion of waveguided modes and SPP modes follows the similar E-k relation of a free travelling photon, where high energy photon state generally has larger kₓ.
The case discussed before doesn’t consider the electroluminescent (EL) spectrum of an emitter, i.e. it assumes the emitter generates photons of all energies in the same intensity. In reality, every emitter has a unique EL spectrum. To achieve the highest outcoupling efficiency, the OLED matrix is tailored for the spectrum of an emitter. Figure 2-2 shows an OLED device using Ir(ppy)₂(acac) as the green emitter, and the EL spectrum of Ir(ppy)₂(acac)⁴⁶. Ir(ppy)₂(acac) emits photons with energy from 450 nm to 700 nm, with the most intensity concentrated around 520 nm. When it is used as a green emitter in an OLED, the photons will couple to each energy level mapped out in Figure 2-1. The result is the mode distribution of Ir(ppy)₂(acac) in the matrix of OLED structure (Figure 2-3).

Setfos integrates the power of the photons that fall in each mode, and use a percentage to represent the ratio of energy in each mode over all the energy emitted by the emitter. From the simulation, 30% of the light emits into air, 24% of the light is trapped in substrate mode, 16% of the light is trapped in the waveguided mode, 26% of the light is trapped in SPP modes. Another 4% of the light is lost due to absorption loss, which is not shown in the mode distribution. The mode distribution tells us there is multiple loss channels in an OLED device.

Setfos is a powerful tool which allow us to modify the OLED structures at will, and foretell how optical properties change with each modification. In the next few paragraphs we will change some of the key layers of OLED structure to gain understanding of the optical cavity. We consider the most typical simple OLED structure, which is a bottom emitting OLED with a glass as substrate, aluminum as the cathode, and ITO as the anode.
Effect of ETL Thickness

ETL is the layer which separates emitter with the reflective metal electrode. For ease of demonstration, the emitter is located at the interface between EML and ETL.

ETL serves three important roles. Firstly, it transports electrons to the EML. The thickness of ETL can affect the device charge balance. Common ETL materials have lower mobility than their HTL counterparts\(^\text{63}\), therefore thick ETL materials are usually unfavorable to achieve balanced charges. In some cases, ETL can be doped with n-type dopant to improve the electron transport.

Secondly, ETL has the strongest effect on which wavelength is supported by the optical cavity. Because photons emitted towards Al is efficiently reflected (over 90%), they can bounce back and interfere with the photons emitted towards ITO. The optical path difference between these two groups of photons are

\[
\Delta = 2 \cdot n_{\text{ETL}} \cdot d_{\text{ETL}},
\]

where \(d_{\text{ETL}}\) is the thickness of ETL. The criterion for constructive interference is

\[
\frac{2\pi}{\lambda} \cdot \Delta + \phi = m \cdot 2\pi,
\]

where \(\phi = \pi\) represents the phase shift of light reflected from the metal surface, \(m\) is an integer (0, ±1, ±2…). This equation tells us that with an increasing thickness of ETL, the constructive interference criterion can be periodically met. We demonstrate this by putting a green emitter (\(\lambda = 520\) nm) in the OLED, and sweep the ETL thickness from 20 nm to 300 nm to observe the change in mode distribution. The result (Figure 2-4) shows sinusoidal change of air mode against ETL thickness. The two air mode peaks correspond to first and second antinode conditions which yield ~30% outcoupling efficiency. The valleys correspond to the destructive interference conditions, and the
outcoupling efficiency can become as low as 5% at these points. This simulation reveals the importance of ETL thickness on the optical cavity. Choosing the right or wrong thickness for ETL can make 5 times difference in device efficiency.

The For first antinode condition \((m = 1)\), the optimal thickness of ETL for constructive interference is

\[
 n_{ETL} d_{ETL} = \frac{\lambda}{4} \quad (2-3)
\]

This equation tells us that for a fixed \(n_{ETL}\) (which is often close to 1.75), the longer the emitter wavelength, the thicker the ETL needs to be to achieve constructive interference. We can demonstrate this with the white emission plot in Setfos (Figure 2-5). This plot is calculated by assuming an emitter that emits in all the wavelength at unity intensity ('white' spectrum). The optical cavity enhances the emitted spectrum at the constructive interference wavelengths. We sweep ETL thickness from 20 nm to 100 nm, and observed enhancement peak shift from \(\lambda = 450\) nm to \(\lambda = 660\) nm, consistent with our theory. The difference in the peak intensity mainly comes from (1) the dispersion of absorbance and reflectance of Al electrode. Lower absorbance and higher reflectance at a certain wavelength will result in higher peak intensity. (2) Loss channels such as SPP mode and waveguided modes. This point will be further discussed below.

Another key role of ETL thickness is determining the coupling strength between the emitter and the SPP mode. The closer the emitter is placed to the metal electrode, the stronger the coupling is\(^{62}\). We plot out the absolute power coupled in to each mode (absolute contribution) against ETL thickness from 20 nm to 80 nm in Figure 2-6. As the thickness of ETL approaches 20 nm, SPP mode shows fast increase in intensity. As a major loss channel, SPP mode can be effectively reduced by increasing ETL thickness.
As a matter of fact, SPP mode can be reduced to <10% in the second antinode condition. This is the main reason the second antinode air mode percentage (30%) is higher than the first antinode air mode (27%).

It should be noted that thicker ETL will inevitably lead to stronger waveguide modes; and any change in the optical cavity will also affect the amount of light coupled into the substrate modes. The outcoupling efficiency is determined by the interplay of air mode (peaks in the constructive interference condition), SPP modes (decreases as ETL thickness increases), waveguided mode (increases as ETL thickness increases) and substrate mode (indirectly affected by the change of optical cavity).

**Effect of HTL Thickness**

The role of HTL can also be divided into electrical and optical. From electrical point of view, HTL thickness affects the transport of holes, which affects the balance of charges. Most HTL materials have much higher hole mobility than their ETL counterparts. A thicker HTL is usually preferred to match the slower transport of electrons.

To explain the optical role of HTL, we can maintain the ETL thickness as constant and sweep the thickness. In this simulation, we continue using green emitter (λ = 520 nm). The thickness of ETL is fixed as 65 nm, which is the optimal thickness from previous simulation. The mode distribution shows a similar sinusoidal change of air mode with increasing HTL thickness (Figure 2-7). However, the air mode peak (27%) and valley (18%) difference is only 1.5 times, compared to the 5 times difference by changing ETL thickness. This indicates HTL also influences the optical cavity, but has a smaller influence than ETL.
We can explain this phenomenon with the cavities formed in an OLED. We’ve mentioned the interference of photons traveling towards the ITO and the photons bounced back from Al cathode determines the optical cavity. However, the two group photons will not just escape the device afterwards. The interfaces between HTL \((n = 1.75)\) and ITO \((n = 1.9)\), ITO \((n = 1.9)\) and glass \((n = 1.5)\), as well as the interface between glass \((n = 1.5)\) and air \((n = 1)\) can all reflect photons back to the OLED structure, which in turn interfere with the emitted light. Therefore, there are multiple cavities in existence in an OLED structure, and the outcoupling efficiency is the interplay of all these cavities.

We plot out the absolute contribution by changing HTL thickness from 20 nm to 500 nm. The power of SPP mode decreases slightly as HTL increases to 100 nm, and remains constant with thicker HTL. This is because as HTL thickness increases, the distribution of emitted power shifts away from the Al cathode, causing a smaller portion of energy being coupled to SPP mode. The increase of HTL thickness also affects the distribution of power in the air mode, substrate mode, and waveguide mode. It can be seen air mode competes with substrate mode in their power. As the HTL thickness increases, the optical cavity is favorable to one of these two modes periodically. This is the result of the cavity formed between air/glass interface and reflective metal. In a similar manner, waveguide mode also competes with substrate mode. This is the result of the cavity formed between glass/ITO interface and reflective metal. The intensity of air mode peak decreases from first antinode to higher order antinode. This is because more energy is distributed to substrate and waveguided modes as HTL becomes thicker.
Optimize both ETL and HTL Thickness

We summarize the effect of optical cavity by sweeping both HTL and ETL from 20 nm to 300 nm. The outcoupling efficiency is plotted in Figure 2-9. Going from first antinode to second antinode by increasing HTL thickness introduces additional waveguided loss, and the efficiency is decreased slightly from 27% to 26%. Going from first antinode to second antinode by increasing ETL thickness can reduce the SPP loss channel, thus increasing the outcoupling efficiency from 27% to 30%. OLED devices commonly don't go to second antinode because the gain of efficiency is marginal compared to the additional material consumption and difficulty of charge balance (ETL doping is mandatory given it already has lower mobility than the HTL).

The aforementioned simulation is based on single emission wavelength. In reality, the OLED spectrum is never a singularity, and always has a spectra power distribution. The actual outcoupling efficiency is the weighted average of all the photons based on the emitter’s spectrum. This requires a great amount of calculation power, which can be achieved with a software like Setfos.
Figure 2-1. A schematic drawing of the mode in an OLED device and its corresponding mode distribution simulated in Setfos

Figure 2-2. An OLED structure using Ir(ppy)$_2$(acac) as the green emitter and the PL spectrum of Ir(ppy)$_2$(acac)
Figure 2-3. Simulated mode distribution of an OLED using Ir(ppy)$_2$(acac) as the green emitter.

Figure 2-4. Schematic drawing of a green OLED device and the simulated mode distribution by changing ETL thickness.
Figure 2-5. The simulated white emission plot by varying ETL thickness from 20 nm to 100 nm.

Figure 2-6. The simulated absolute contribution of each mode by sweeping ETL thickness from 20 nm to 100 nm.
Figure 2-7. Schematic drawing of a green OLED device and the simulated mode distribution by changing HTL thickness.

Figure 2-8. The simulated absolute contribution of each mode by sweeping HTL thickness from 20 nm to 500 nm.
Figure 2-9. The simulated air mode intensity by changing ETL and HTL thickness to fulfill 1\textsuperscript{st}, 2\textsuperscript{nd} and 3\textsuperscript{rd} antinode conditions.
CHAPTER 3
OLED LIGHT EXTRACTION WITH CORRUGATION

Light Extraction in OLEDs

Based on the previous simulation, we can see only a quarter of the light generated by the emitter can escape the device. A significant portion of the light is trapped in SPP mode, substrate mode and waveguided modes. Many light extraction techniques have been applied to recover the trapped lights in an OLED\textsuperscript{5-6, 12, 61, 66}. Each of the techniques targets at minimizing or extracting light from one or more of the loss channels (Figure 3-1). The feasibility of these techniques is examined by their EQE improvement amount, cost and compatibility with current OLED manufacturing methods. From optical point of view, we categorize these light extraction techniques by the trapped mode they target at. Sub-anode grids using high index TiO\textsubscript{2} and low index SiO\textsubscript{2} is effective in extracting the waveguided modes\textsuperscript{10}; corrugation on both bottom anode and top electrode is effective in extracting both waveguided mode and SPP modes\textsuperscript{2, 5, 60}; Half-ball lens and micro lens arrays outside the substrate can remove the total internal reflection at the glass/air interface, and is effective in extracting substrate modes\textsuperscript{5}; Porous substrate which includes air voids can effectively scatter and extract substrate mode. With the combination of one or a few of the light extraction techniques, the OLED device EQE has been improved from 20\% to 50-60\%\textsuperscript{12, 67}.

This research focuses on corrugation, which is effective in extracting both waveguided mode and SPP mode. In this chapter, we will go through how corrugation extracts trapped modes and what are the basic requirements for corrugation light extraction.
Mode Extraction with Corrugation

When a ray of light is incident on a grating, it will be diffracted and create several rays of diffracted light. The diffraction equation is

\[ k \sin \theta_i + m \cdot k_G = k \sin \theta_d , \quad (3-1) \]

where \( k = \frac{2\pi}{\lambda} \) is the wave-vector of the incident light (and the diffracted light), \( \theta_i \) is the angle of the incident light, \( \theta_d \) is the angle of the diffracted light, \( k_G \) is the diffraction vector of the grating, \( m \) is a non-zero integer (\( m = \pm1, \pm2 \ldots \)). Each \( m \) value corresponds to the order of diffraction. In a common diffraction grating, diffraction light intensity is the strongest in the first order diffraction, and decreases as the order increases. In the following chapters, only the first order diffraction is considered.

In an OLED device, \( k \sin \theta \) is generalized to the in-plane wave-vector \( k_x \) of a certain mode. This is especially useful when considering the diffraction of SPP mode, since \( k_x \) of a SPP mode is larger than \( k \), making \( \theta \) non-exist. As we discussed before, \( k_x \) is the key parameter which determines whether a photon can escape the device. Only photons with \( 0 \leq k_x < k_0 \) can contribute to the air mode. Waveguided mode and SPP modes have a defined \( k_x \) vector that is greater than \( k_0 \), causing them to be trapped in the OLED devices.

While mode of the corrugation patterns reported for OLED extraction has a defined pitch, or a narrow range of pitch values, all corrugation patterns can be Fourier transformed to reveal its distribution of pitch values. In the following discussion, we assume a corrugation with a pitch of \( \Lambda \).

Corrugation extracts waveguide and SPP modes by Bragg diffraction. Its grating vector \( k_G \) is calculated as
\[ k_G = \frac{2\pi}{\Lambda}, \quad (3-2) \]

where \( \Lambda \) is the pitch of the corrugation. When waveguided mode and SPP mode interact with the corrugation, the first order diffraction causes \( k_x \) to be reduced by \( k_G \),

\[ k_d = k_x - k_G. \quad (3-3) \]

The case of \( k_d = k_x + k_G \) is energetically not favored because there is generally no mode supporting larger \( k \) values than the trapped mode, so light is unlikely to be diffracted to larger \( k \) values.

When \( |k_d| \) is smaller than \( k_0 \), the photon is diffracted out of the device and contributes to the air mode. This is the theoretical ground of light extraction with corrugation. In the simulation of a green OLED device, at the peak emission wavelength \( \lambda = 520 \text{ nm} \), air line edge is at 12 \( \mu\text{m}^{-1} \), SPP is located around 23 \( \mu\text{m}^{-1} \). For a corrugation to effectively extract SPP mode at 520 nm, the grating vector needs to satisfy

\[ 0 \leq |k_{\text{SPP}} - k_G| < k_0. \quad (3-4) \]

Solving this equation gives us the range of \( k_G \) to be between 11 \( \mu\text{m}^{-1} \) and 35 \( \mu\text{m}^{-1} \), corresponding to a range of corrugation pitch between 180 nm and 570 nm. Note that \( k_G \) is independent of the emission wavelength \( \lambda \), which means corrugation diffraction only shifts the mode along the \( k_x \) axis, while preserving the dispersion relation of shifted mode.

The same can be calculated for waveguided mode extraction. Because waveguided modes have smaller \( k_x \) values, a smaller \( k_G \) is needed to diffract them across air line, therefore, larger pitch is sufficient for waveguided mode extraction. In the case of \( \lambda = \)
520 nm, corrugation pitch between 220 nm and 1.2 μm can extract waveguided modes. Air mode and substrate mode are not affected by the grating as much as waveguided mode and SPP mode. Because air mode and substrate modes are incoherent. The diffraction from corrugation is less effective on incoherent modes because light doesn’t have a fixed phase relation which causes interference effect weaker than in coherent modes (i.e. waveguide and SPP modes).

The amount of light extraction with corrugation is determined by the amount of trapped light and the diffraction efficiency of corrugation. Generally speaking, a larger corrugation depth results in more efficient extraction. However, larger corrugation depth also affects the electrical field distribution in an OLED, which results in the current going through the thinnest parts between two electrodes. In addition, large corrugation depth makes the device prone to shorting problems, severely reducing device stability.

**2D Mode Distribution and Diffraction**

So far, we only considered the distribution of modes along one axis \( k_x \). In practice, the corrugated substrate always carries a 2D pattern, which diffracts light inside the \( k_x-k_y \) plane. For a device fabricated on a planar substrate, its optical properties are center symmetrical. This means the mode distribution of a planar device is identical for the -\( k \) and +\( k \) axes. Using Setfos simulation, we can calculate the mode distribution of a green device in the +\( k \) axis, and use mirror image to reproduce the mode distribution in the -\( k \) axis (Figure 3-4).

Because the planar device is isotropic in the \( k_x-k_y \) plane, the mode distribution in the \( k \) axis shown in Figure 3-4 is identical for any device orientation \( \varphi \). This means we can use the mode distribution calculated in +\( k_x \) axis to reproduce the mode distribution in the entire \( k_x-k_y \) plane. We demonstrate this by taking the mode intensity at the emission
peak of $\lambda = 520$ nm (Figure 3-5). From the mode intensity plot, we can see the air mode and substrate modes are continuous, while waveguided modes are discrete and narrow. By projecting the mode intensity from a random $k$ axis to every direction in the $k_x$-$k_y$ plane, we obtain the 2D mode intensity for the simulated device at $\lambda = 520$ nm. In this 2D plot, the air mode is a disk. The center of the air mode is the strongest, and towards the edge the mode intensity becomes weaker. In 2D mode plot, the air line edge becomes a circle, which has a radius of $k_0$. The substrate mode is a ring outside the air mode. TM waveguided mode, TE waveguided mode and SPP mode are three concentric circles, representing the strong localization of energy along the perimeter. The emission intensity of an OLED device inside the air mode is called its far field emission. For a planar device, its far field emission is uniform and featureless. The emission intensity close to the air mode edge is weaker than the center of air mode. This is due to the reduction of transmittance from glass to air at large angles.

Now let’s consider a simple 2D corrugation pattern, which is a 1D grating along the $x$ axis. The diffraction of this corrugated substrate is along the $x$ axis, with diffraction wavevectors $k_{G1} = \frac{2\pi}{\Lambda}$ and $k_{G2} = -\frac{2\pi}{\Lambda}$. When the same OLED device mentioned before is fabricated on this corrugated substrate, the trapped modes will be diffracted by the corrugation, but only along the $x$-axis. We consider the air mode and the SPP mode (Figure 3-6). The air mode is represented by its edge $k = k_0$, SPP mode is represented by $k = k_{SPP}$. The SPP mode lies completely outside the air mode, indicating it cannot emit. When SPP interacts with the corrugation, the SPP mode forms two diffracted SPP modes. Diffracted SPP$_1$ is shifted from the SPP circle by a vector of $k_{G1}$, while diffracted SPP$_2$ is shifted from the SPP circle by a vector of $k_{G2}$. Now the two diffracted modes
each has an arch inside the air mode. This part of energy is considered extracted from
the device.

**Single Photonic Crystal Patterns**

In this research, we use photonic crystals as the corrugation pattern. Photonic
crystals are periodic optical nanostructure that can interact with photons, in a similar
manner that ionic lattices in solid materials interact with electrons. Photonic crystals
usually have a feature scale similar to or smaller than the wavelength of the photons
they interact with. For OLED application, the wavelength of the photons is between 400
nm to 700 nm. The photonic crystals used in OLED research often have a pitch from
200 nm to 2 μm\(^{68-70}\).

In this research, two categories of photonic crystal patterns are studied. First
category is the single photonic crystals. These photonic crystals have both short-range
and long-range orders. One sample of the single photonic crystals is shown in (Figure
3-7). This photonic crystal has a hexagonal lattice structure, rod-shaped unit, with a
unit-to-unit distance of 400 nm. Similar to how an electron interacts with an atomic
lattice, when light interact with this photonic crystal, it is actually interacting with the
arrays of lattice units. Therefore, the pitch of this photonic crystal is the distance
between the two closest arrays of lattice units:

\[ \Lambda = d \cdot \sin \frac{\pi}{3} = \frac{\sqrt{3}}{2} d = 346 \text{ nm} \ . \]

The diffraction vector of this photonic crystal is

\[ k_o = \frac{2\pi}{\Lambda} = 18.1 \mu \text{m}^{-1} \ . \]
To study the diffraction properties of this photonic crystal, we draw its lattice points in the reciprocal space (i.e. a 2D Ewald construction) in Figure 3-7. Here we only plot out the origin point (P₀) and the first order diffraction lattice points (D₁ to D₆). The reciprocal space has the same hexagonal symmetry as the real space, but is rotated 30°. The six adjacent lattice points next to the origin point represents the six directions of first order diffraction (six directions of \( \vec{k}_a \)). The direction of \( \vec{k}_a \) in the reciprocal space corresponds to the direction of one optical axis in the real space, and the angle \( \varphi \) in the reciprocal space corresponds to the substrate orientation angle \( \varphi \) in the real space.

We consider a photon with an in-plane wave-vector of \( \vec{k} \). Assume this photon is trapped in an OLED device in either waveguided mode or SPP mode, so that \( \vec{k} > \vec{k}_0 \). The air mode edge is represented as a circle with a radius of \( k_0 \). When this photon is incident on the photonic crystal, it can be diffracted by any of the six diffraction vectors. The resulting diffracted wave-vector becomes

\[
\vec{k}_{\text{diff}} = \vec{k} + \vec{k}_a
\]

(3-6)

Only one of the six \( \vec{k}_{\text{diff}} \) is shown in Figure 3-7. Of the six diffracted light, only light diffracted by D₁ ends up inside the air cone. To put it another way, only light diffracted by D₁ satisfies the condition \( \left| \vec{k}_{\text{diff}} \right| < k_0 \).

Previously we have mentioned waveguided modes and SPP mode are concentric circles in 2D mode distribution. So far, we only considered the diffraction of trapped mode in one direction. To study the diffraction of trapped modes in all directions, we need to shift the waveguided mode or SPP mode circle according to the six \( \vec{k}_a \) vectors. For simplicity, let’s consider only the SPP mode. The 2D mode plot is shown in Figure
The photonic crystal diffracts the SPP mode to 6 directions, forming 6 diffracted SPP modes. Each mode is shifted along one of the optical axes by a vector of $k_G$. The diffracted SPP modes each has an arch inside the air mode. All the arches combined form a lotus flower pattern in the far field emission.

The same process can be done for the two waveguided modes (Figure 3-9). Taking TM waveguided mode as example. Because it has a smaller $k$ vector than the SPP mode, the TM waveguided mode circle is closer to the air mode. TM waveguided mode is also diffracted by the photonic crystal to 6 directions, forming 6 diffracted TM waveguided modes. The arches of these diffracted modes form a lotus flower pattern in the far field emission, but is closer to the center of the air mode.

The light extraction effect of the corrugation is calculated by considering the diffraction of all the trapped modes (including TM waveguided mode, TE waveguided mode and SPP mode), and calculating the amount of diffracted energy distributed in the air mode. The diffraction efficiency of the corrugation determines how much energy is funneled into the diffracted modes.

**Poly Photonic Crystal Patterns**

We also studied the diffraction of poly photonic crystal patterns. Compared to the single photonic crystal, poly photonic crystal only has short ranged order, and is random in the long range. Figure 3-10 shows the SEM image of the surface of the poly crystal substrate. In Region 1 and Region 2, the lattice is hexagonally symmetrical, and the unit-to-unit distance is 400 nm. Therefore, the short ranged pitches in both regions are 346 nm. However, the optical axis orientations are different in Region 1 and Region 2 ($\phi_1 \neq \phi_2$). The lattice orientation transits from Region 1 to Region 2, forming a transitional area in between. This area can be considered as the ‘grain boundary’.
Depending on the processing method, the ‘grain size’ of studied poly crystal is estimated to be 10 μm. On an OLED pixel (2 mm by 2 mm), there are about 40,000 grains, each has a random grain orientation. The overall diffraction effect by the corrugation is uniform in any directions. In reciprocal space, the first order diffraction lattice points are continuous and form a ring, indicating any diffraction direction is equal in intensity for a poly crystal. For instance, both $k_G$ and $k_G'$ can diffract $k$ inside the air mode (D and D'). There are infinite numbers of possible points between D and D' that satisfy the light extraction criterion. Random diffraction orientation is the most significant difference between single and poly photonic crystals.

Another difference between single and poly photonic crystal is the distribution of $k_G$. In single photonic crystals, the entire substrate is one grain, so the pitch $\Lambda$ is consistent throughout the entire substrates. In poly photonic crystals, there are always some crystal defects in the areas along the grain boundaries. The defects include dislocation and vacancies, both of which affect the distance between lattice units. Therefore, although the pitch $\Lambda$ is well defined in each grain, the defects cause the $k_G$ distribution to be wider than in the single crystal. This is manifested as a widened diffraction ring in the reciprocal space. The widening of $k_G$ is dependent on the processing method. Despite the defects, the dominant corrugation pitch $\Lambda$ determines the most intense diffraction to occur at $k_G = \frac{2\pi}{\Lambda}$.

We now investigate the mode diffraction in an OLED device using poly crystal corrugation. For simplicity, only SPP mode is considered. The 2D mode plot is shown in Figure 3-11. SPP circle is diffracted by the poly crystal in all directions, and the shift amount is $k_G$. Therefore, the centers of the diffracted SPP circles are along a circle with
a radius of $k_G$. Each diffracted SPP circle has an arch inside the air mode. The integration of all the extracted features form a ring in the air mode. The inner rim of the ring is stronger in intensity, the radius is $|k_{SPP} - k_G|$; the outer rim of the ring extends to the edge of the air mode, and is weaker as the radius increases. When taking consideration of the distribution of $k_G$, the inner rim of the ring is not perfectly defined.

We can also understand the pattern in an intuitive way. Take the lotus flower pattern in the single photonic crystal (for a single grain orientation $\varphi$), and spin it by the center of air mode (integrate over all the $\varphi$ angles). The resulting pattern is exactly the case for a poly photonic crystal. The mode intensity along the radius is the strongest when close to the center of air mode. The closest point to the center of air mode is $|k_{SPP} - k_G|$. In this sense, the extraction efficiency of a single and poly crystal with the same profile and pitch should be identical. The only difference should come from the defects in poly crystals which changes the distribution of $k_G$. 
Figure 3-1. A schematic drawing showing some of the light extraction methods used to improve OLED outcoupling efficiency.

Figure 3-2. Light extraction with corrugation. The mode distribution plot shows the $k_x$ of trapped modes and the effect of diffraction on reducing their $k_x$. 
Figure 3-3. Mode distribution of a planar device including both -k and +k directions. The dashed line represents the emission peak (λ = 520 nm) of the green emitter Ir(ppy)2(acac).
Figure 3-4. Mode intensity of a green OLED device at $\lambda = 520$ nm and its projection in the $k_x$-$k_y$ plane.
Figure 3-5. Schematic drawing of a 1D grating and the diffraction of SPP modes.

Figure 3-6. SEM image of a single photonic crystal pattern and the Ewald construction of the pattern (only showing first order diffraction).
Figure 3-7. Diffraction of the SPP mode by a single crystal corrugation and its far field emission pattern.

Figure 3-8. Diffraction of the waveguided mode by a single crystal corrugation and its far field emission pattern.
Figure 3-9. SEM image of a poly photonic crystal pattern and its Ewald construction (only showing first order diffraction).

Figure 3-10. Diffraction of the waveguided mode by a poly crystal corrugation and its far field emission pattern.
Setfos simulation can accurately determine the mode distribution of a planar device where each layer is homogeneous. But it cannot deal with more complicated OLED with nano-structures, such as nano-rods, nano-particles, roughening layer or corrugation. This makes it very difficult to determine the origin of extracted light, and quantify the effectiveness of light extraction techniques. Angle resolved electroluminescence spectroscopy (ARES) can serve the purpose, and create new depth for optical study of OLED devices.

**Setup for ARES Measurement**

To experimentally determine the optical properties of any OLED, it’s important to plot out its air mode. This is especially useful when studying light extraction with corrugation. As mentioned before, when waveguided mode and SPP mode are extracted by the corrugation, they are shifted to the air mode while preserving the dispersion relation. Because air mode is continuous and featureless, we should be able to observe the outstanding extracted mode by the locally stronger intensity and the dispersion relation of diffraction feature.

The setup for ARES measurement is shown in Figure 4-1. It’s composed of a rotary stage, an OLED sample holder, an optical fiber and a spectrometer. The sample holder carrying the OLED device is mounted on the rotary stage. An optical fiber is positioned about 12 cm away from the sample holder, pointing at the OLED pixel to collect emitted light. The fiber is connected to the spectrometer which processes the light signal and plot out the EL spectrum of measured OLED device. The axis of the rotary stage and
the optical fiber is aligned with a laser pointer, so that when the rotary stage rotates, the optical fiber always points at the OLED pixel.

We explain the working principle of this setup as follows: the OLED device is installed on the sample holder at a certain device orientation (φ). This is useful when measuring devices fabricated on corrugated substrates with anisotropic substrate. The rotary stage controls the facing angle (θ) of the OLED device. The angle resolution of the rotary stage is 0.9°. When rotating the device, the optical fiber collects the photons travelling along the facing angle θ. The signal is collected by the spectrometer which generates an electroluminescent (EL) spectrum at given facing angle θ.

To characterize the air mode of an OLED, we need to sweep the device facing angle from -90° to 90° (Figure 4-2). This range corresponds to $k_x = k_0 \sin(-90°) = -k_0$ and $k_x = k_0 \sin(90°) = k_0$, thus mapping out the photons within the air lines. The wavelength limits of the spectrometer determine the range of photon energies. With Ocean Optics HR 4000, the EL spectrum is measured from 195 nm to 1124 nm, which translates to 1.1 eV to 6.4 eV. This range covers the visible wavelength range 400 nm to 700 nm.

The measured EL spectra are processed to mapped out the air mode. Each data point in the EL spectrum represents a group of photons. Figure 4-3 demonstrates an EL spectrum collected at $θ = 30°$. The red dot represents the EL intensity at 540 nm. This data point translates to a point in the air mode with photon energy of $E = \frac{hc}{\lambda} = 2.29$ eV, and in-plane wave-vector $k_x = \frac{2\pi}{\lambda} \sin(30°) = 6.04$ μm$^{-1}$. The mode intensity is calculated by multiplying the EL intensity by a factor of $1/\cosθ$, which takes into consideration of the projected area of the device at θ angle. The resulting air mode intensity is calibrated to per unit area.
By doing this for all the EL spectrum collected from \(-90^\circ\) to \(90^\circ\), we obtain the air mode of the measured OLED device at device orientation \(\phi\). In most cases, the substrate is center symmetrical, meaning the number of photons travelling in the angle \(\theta\) and \(-\theta\) are identical. Therefore, it's often sufficient to only measure air mode from \(0^\circ\) to \(90^\circ\), and use symmetry to mirror the other half of air mode.

In some cases, we need to study the polarization of light. This can be achieved by inserting a polarizer between the device and the optical fiber. When the optical axis of the polarizer is along the reference plane, the optical fiber collects P polarized light. When the optical axis is perpendicular to the reference plane, the optical fiber collects S polarized light.

**Performance of Planar Device**

An OLED device is fabricated on a planar glass substrate. The OLED device structure is shown in Figure 4-4. ITO is used as the anode, and Al is used as the cathode. 10 nm MoO_x is used as the HIL, 40 nm TAPC is used as the HTL, 20 nm CBP doped with 7% Ir(ppy)_2(acac) is used as the emitter, 10 nm B3PYMPM is used as the HBL, 30 nm Alq3 doped with 10% Cs_2CO_3 is used as the ETL, 2 nm Cs_2CO_3 is used as the EIL. The thickness of the glass substrate is 1 mm. The pixel area of the device is 2 mm by 2 mm. A picture of the pixel driven at 1.25 mA/cm^2 is also shown. The pixel is uniform and has no haze.

The performance of the planar OLED device is characterized by its J-V curve, current efficiency, EQE and forward EL spectrum (Figure 4-5). The J-V curve shows a turn-on voltage of 2.4 V, similar to the bandgap of the green emitter Ir(ppy)_2(acac). Below the turn-on voltage, the leakage current level is below 50 nA/cm^2. The device has a current efficiency of 91 cd/A from 10 to 100 cd/m^2. Its EQE is peaked at 27%. The EL
spectrum shows a narrow peak at 520 nm, and a shoulder at 550 nm. Simulation of the planar device indicates a peak air mode of 30%. Considering \( \text{Ir(ppy)}_2(\text{acac}) \) has a quantum yield of 95%, the EQE of an optimized device should be 28.5%. The discrepancy between the measured EQE and simulated EQE comes from (1) variation of layer thickness and refractive index. This planar device uses sputtered ITO, which has slightly lower transmittance and higher refractive index than the commercial ITO. Also, the calibration of layer evaporation can be slightly off, causing the optical stack thickness to be different from the optimized values. (2) The charge balance of the device can be slightly off. The 1.5% difference could come from 95% charge balance instead of 100%. (3) The difference is within the EQE measurement error margin. Nonetheless, this planar device demonstrates high efficiency close to the optimal value.

**ARES Study of Planar Device**

We used ARES setup to measure the EL spectra of said planar device from 0 to 90°, and plot them together. The result shows a homogeneously decreasing intensity of the entire EL intensity from 0 to 90°. We also normalize the EL spectra in Figure 4-6 to show see the distortion of spectrum from 0 to 70° (higher angle spectra are not included due to the larger noise to signal ratio). From the normalized spectra, we can see a small distortion of the spectrum, mainly due to the reduction of shoulder and tail intensity longer than 550 nm. This is because at larger angles, the effective cavity length is reduced, therefore, the peak of cavity modes blue shifts. We can demonstrate this effect in Setfos by sweeping the white emission at different viewing angles (θ). The result (Figure 4-7) shows a blue shift of cavity mode peak from 550 nm to 475 nm. This means at larger θ, the cavity supports shorter wavelength, which causes the emission spectrum to blue shift. If we take consideration of the cavity mode, and backtrack the EL spectra
by dividing the white emission, we obtain the so-called ‘intrinsic’ EL spectrum. The intrinsic EL spectra calculated at 0 to 70° overlap with each other, which shows a good agreement between the simulation and experiment. The intrinsic spectrum is determined by the emitter molecule, host-emitter interaction, doping concentration, and is independent on the rest of the OLED device structure.

We plot out the air mode of the planar device based on the measured EL spectra, and compare it with the Setfos simulation result (Figure 4-8). The planar device air mode is continuous and featureless. At the emitter intensity peak λ = 520 nm, the intensity of the air mode is the strongest. The experiment and simulation agree very well, confirming the validity of the simulation method.
Figure 4-1. The setup for ARES measurement.

Figure 4-2. Schematic drawing of the process of measuring an OLED device with ARES setup.
Figure 4-3. Using EL spectrum to determine the intensity, photon energy and in-plane wavevector of a photon group.

Figure 4-4. OLED structure of the planar device and a photo of the pixel driven at 1.25 mA/cm$^2$. 
Figure 4-5. Performance of a planar OLED device (a) J-V curve, (b) current efficiency plot, (c) EQE plot, (d) forward EL spectrum
Figure 4-6. (Left) EL spectra of a planar OLED at different viewing angles. (Right) The normalized EL spectra of the same device showing a blue shift of the spectrum due to shift of cavity mode.

Figure 4-7. (Left) White emission (cavity mode) of the OLED device at viewing angles from 0 to 90°. (Right) Normalized EL spectra divided by the cavity mode curve at each angle showing identical green emission spectrum, the ‘intrinsic’ EL emission spectrum.
Figure 4-8. Comparison of the (left) measured and (right) simulated air mode of a planar OLED.
CHAPTER 5
ARES STUDY OF CORRUGATED OLED DEVICES FABRICATED ON SINGLE AND POLY PHOTONIC CRYSTAL SUBSTRATES

Preparation of Corrugated Substrate

Corrugation is a nano-structure with uneven surface profile. The nano-structure has several key parameters, including pitch, depth and shape. There are a few ways to generate a corrugated profile, including interference lithography, holographic lithography, thermal induced self-buckling, UV induced self-buckling, and monolayer nano-sphere patterning. After a corrugated structure is formed, it can be used as a master mold. The corrugation pattern can be transferred from a mold to a substrate through nano-imprinting. The substrate is then used to fabricate OLED devices, so that every layer sputtered or evaporated on the corrugated substrate follows the same surface profile.

In this research, some of the corrugation molds are provided by MicroContinuum, the others fabricated through mono-layer silica sphere nano-patterning. The details for the latter is explained elsewhere. With the master molds ready, corrugated substrates are prepared by nano-imprinting. The fabrication of a corrugated OLED is described in Figure 5-1. First, a PDMS: initiator mixture is prepared at raito of 9:1. Then the mixture is drop-casted on the corrugated mold, and goes through thermal annealing at 65° for two hours to crosslink. After thoroughly crosslinking, the PDMS stamp is peeled off and set aside. A glass substrate is cleaned with standard acetone and isopropanol solvent process. Then the glass substrate is treated with 365 nm UV light (14 mW/cm²) for 4 minutes to reduce surface energy. Then a small amount of optical adhesive NOA 81 (which has a matching refractive index with glass) is drop-casted on the planar glass substrate. The PDMS stamp carrying the negative form of corrugated pattern is then
pressed onto the optical adhesive. The substrate is then treated with UV light for 4
minutes to cross-link the adhesive. Then the pattern is preserved. Finally, the PDMS
stamp is removed and the corrugated substrate is ready.

The as prepared substrate usually has a large corrugation depth (over 120 nm)
which can cause leakage current in the device. Proper planarization with PEDOT: PSS
done with spin-coating is needed to control the corrugation depth. After planarization the
substrate is put into sputter for ITO sputtering. Then the substrate is put into a vacuum
chamber to evaporate the rest of the OLED structure.

**ARES Study of Single Photonic OLED**

We fabricate an OLED device on corrugated substrate carrying a single photonic
crystal pattern (abbreviated as single crystal device). The pattern has a hexagonal
lattice structure, with a pitch of 346 nm (unit-to-unit distance of 400 nm). The depth of
the corrugation is around 90 nm. The OLED structure is the same as the planar device
discussed before, so the two devices make a fair comparison to show the effect of using
corrugated substrate.

Figure 5-2 shows an operating pixel of the corrugated device. The pixel is uniform
with small amount of haze, and we can also see noticeable glares in 6 directions. The
performance of the device is shown in Figure 5-3. We use the planar device with the
same OLED structure as a reference. The corrugated device has a turn-on voltage at
2.4 V, same as the planar device. The leakage current below turn-on voltage is around
200 nA/cm², higher than the planar device. The corrugated device shows a peak current
efficiency of 120 cd/A, 32% higher than the planar device. The measured EQE is 33%,
22% higher than the planar device. We notice some interesting difference between the
corrugated device and the planar device. The corrugated device has a similar
background as the planar device, with additional ‘bumps’ around 540 nm, 590 nm and 630 nm. We can conclude these bumps are unique features at the forward viewing angle by comparing the EL spectra at viewing angles from 0 to 90° (Figure 5-4). As the viewing angle increases, we can see the normal reduction of spectrum intensity, as well as the shift of said ‘bumps’ in different wavelengths.

We use the measured angle dependent EL spectra and plot out the air mode of the corrugated device in Figure 5-5. The single crystal device shows a similar background as the planar device air mode, with additional features (the intensity scale of the corrugated device is larger than the planar device to accommodate for the features). These features look like isolated dots in the emission peak λ = 520 nm, but are actually continuous linear features across every photon energy within the emitter EL spectrum (1.95 eV to 2.55 eV). Based on our discussion before, corrugation extracts waveguided and SPP modes. Both waveguided and SPP modes are narrow and line-shaped, when they are diffracted into the air mode, the diffracted modes should stand out from the featureless air modes. However, because both air mode and diffracted modes have a similar intensity background determined by the emitter EL spectrum, it’s difficult to distinguish the linear modes.

To solve this issue, we can divide the intensity of air mode by the EL spectrum, thus acquiring the so-called ‘normalized air mode’ (Figure 5-6). This process removes the information of the emitter EL spectrum, and reveals the diffracted mode line-shape which is only dependent on (1) the OLED optical cavity and (2) the corrugation. From the normalized air mode we can now clearly see the dot shaped features are indeed linear features which extend across the entire emission spectrum of the emitter. The
linear features in the normalized air mode tend to intercept on several photon energies in the normal direction \((k = 0, \theta = 0^\circ)\). This is the result of the symmetry of \(+k\) and \(-k\) directions for both the device mode distribution and the hexagonal corrugation pattern. To better demonstrate this symmetry, we use a mirrored image to show the normalized air mode across the entire \(180^\circ\) viewing angle (Figure 5-7). Now we can see the continuous linear features of waveguided modes and SPP mode, and understand how they intercept in the same energy in the center of air mode, and continue extending. We can also see how normalization eliminates the information of the emitter, revealing the true density of modes. The features have consistent intensity across photon energies, as is dictated by the Setfos simulation. In the center of air mode, the diffracted modes from \(-k\) and \(+k\) region intercept and form local intensity peaks.

We can assign these diffraction features to the extracted waveguided and SPP modes. The details will be explained in the later sessions. The strongest 3 set of features come from SPP modes, which intercept at around 630 nm in the center. TM waveguided mode also has 3 set features, which are weaker in intensity and intercept at 550 nm. TE waveguided mode is the weakest among the features. Only one set of TE waveguided mode features can be distinguished from the air mode background, which intercept at around 590 nm. This confirms the ‘bumps’ in the device forward EL spectrum come from extracted modes, including TM, TE waveguided modes and SPP modes. Note some of these diffraction features are close to linear-shape, while some are curved.
Assignment of Diffraction Features to Trapped Modes

So far, we have seen the diffraction features in the air mode. Now we will explain how these features are assigned to each mode.

An OLED device has a 3D structure (Figure 5-8). For a planar device, the x-y plane is isotropic. This means if we take a cross-section in any plane normal to the x-y plane, the mode distribution is identical. In Figure 5-7, one slice of such plane is shown, revealing the air mode, one of the waveguided modes and SPP modes of which the in-plane wave-vector $k$ is inside the plane. We concluded for a single photon energy, the air mode is a disk, waveguided modes and SPP mode are concentric circles. When we consider the dispersion of each mode by the photon energies, the mode distribution in the $k_x$-$k_y$-$E$ space becomes cone-shaped. For example, the air mode edge is defined by $k = k_0 = \frac{E}{\hbar c}$, so in a polar plot, the k-E relation is strictly cone-shaped. Sometimes the air mode is called an ‘air cone’ for this exact reason. For waveguide modes and SPP mode, the relation between $k$ and $E$ is not exactly linear, but are not too far off in the visible region. For ease of demonstration, we assume they are also linear.

Now let’s consider the diffraction of SPP modes by the corrugation. First, we assume the diffraction vector $k_G$ is along the $k_x$ axis (Figure 5-9). The SPP cone is shifted along the $+k_x$ and $-k_x$ directions to form two diffracted modes. We now analyze the diffraction pattern from two perspectives. From the top down view ($k_x$-$k_y$ plane) in Figure 5-10, we can see the same far field emission pattern we discussed about earlier. Two diffracted SPP circles each has an arch inside the air cone. From the side view ($k_x$-$E$ plane), the two branches of SPP modes intercept in the center of air cone at $E_0$, the two branches of crossing have the exact same dispersion relation as the original SPP branches.
In the next step, we consider the case where \( k_G \) is in an arbitrary direction (Figure 5-11). Because the direction of \( k_G \) is relative to the \( k\)-\( E \) plane we choose, we only need to look at the mode distribution in one \( k\)-\( E \) plane. In this case, we observe the mode distribution in the \( k_x\)-\( E \) plane, which is labeled green in Figure 5-12. Assume the direction of \( k_G \) is \( \phi \) from the \( k_x \) axis, SPP modes shift in two directions and form two diffracted modes. When we observe the mode distribution in the \( k_x\)-\( E \) plane, we cut through the two diffracted SPP surfaces. We can prove this using geometry knowledge: when we cut a cone vertically from a point that’s not its geometric center, we obtain a hyperbola curve. In the \( k_x\)-\( E \) plane, we see two symmetrical hyperbola curves that correspond to the two diffracted SPP modes by two arbitrary angles. Due to symmetry, the two curves also intercept in the normal direction at the same photon energy \( E_0 \). The equations of the hyperbola curves are dependent on both \( \phi \) and the magnitude of \( k_G \).

When \( k_G \sin \phi \) is small (\( \phi \) is close to 0°), the hyperbola curves approach linear shapes; when \( k_G \sin \phi \) is big (\( \phi \) is close to 90°), the hyperbola curves have large curvature radius close to the center of air cone.

We have discussed the mode distribution of any arbitrary diffraction directions. Now we can apply this conclusion to the case of hexagonal single photonic crystal diffraction. For any device orientation \( \phi \), the diffraction occurs in 6 directions: \( \phi, \phi + \frac{1}{3}\pi, \phi + \frac{2}{3}\pi \), and the opposite directions \( -\phi, -\phi - \frac{1}{3}\pi \) and \( -\phi - \frac{2}{3}\pi \). The diffracted modes from these directions have different shapes, but will intercept at the same photon energy \( E_0 \) in the forward viewing angle. For each mode (TM waveguide mode, TE waveguide mode and
SPP mode), $E_0$ can be calculated with the dispersion of said mode and $k_0$ by considering diffraction along the $k_x$ axis.

**Polarized Normalized Air Mode and Simulation**

The ARES measurement provides a lot of information regarding the photonic crystal and the OLED device. In the previous measurement, $\phi$ angle is selected at random. Mathematically we can find a fit of the hyperbola curves of each mode and derive the actual $\phi$ angle. Although this is under the hypothesis that the waveguide modes and SPP mode are close to linear-shaped within the photon energies discussed. A more precise way is to use the Setfos simulation and obtain the exact dispersion of waveguided and SPP modes. In addition, given that TM waveguide mode and SPP mode are P polarized light, and TE waveguided mode is S polarized light, we can use a polarizer to measure the polarized mode intensity to differentiate light of different polarization, and quantify the diffraction efficiency more precisely.

Here we show the measured mode distribution of a single crystal OLED at two arbitrary device orientations (Figure 5-13, 5-14). A polarizer is used to differentiate S and P polarized lights. We compare the measured result with the mode distribution (Figure 5-15, 5-16). obtained from Setfos simulation done by my colleague Cheng Peng. From the simulation result, we confirm the device orientation is $\varphi_1 = 5^\circ$ and $\varphi_2 = 25^\circ$ for the two cases. The simulation results show excellent agreement with the measurement, revealing the origin of the three set of diffracted lights come from TM waveguided mode, TE waveguided mode and SPP mode.
At $\phi_1 = 5^\circ$, the diffraction directions are $\pm 5^\circ$, $\pm 65^\circ$ and $\pm 125^\circ$ respectively, the three set of diffracted curves are linear shaped in the center of air cone; at $\phi_2 = 25^\circ$, the diffraction directions are $\pm 25^\circ$, $\pm 85^\circ$ and $\pm 145^\circ$. The set of curves at $\pm 85^\circ$ have large curvature radius in the normal direction. This can be easily observed in the normalized plot. From $0^\circ$ to $\pm 60^\circ$, the diffraction peaks around 540 nm, 580 nm and 630 nm remain strong. This observation is important for application where diffraction is needed within a wide range of angles.

We now analyze the polarization properties of each mode. The polarization of a wave is in reference to its plane of wave propagation. This is usually different from the observation plane ($k_x$-E) we choose. Taking SPP mode as an example: SPP mode is strictly P polarized. When SPP light is diffracted by $k_g$ along $k_x$ axis ($\phi = 0^\circ$), its polarization is unchanged. This is reason we see a strong SPP features in ($\phi_1=5^\circ$, P light), which comes from diffraction direction $\phi = 5^\circ$. However, when the SPP light is diffracted perpendicular to $k_x$ axis ($\phi = 90^\circ$), its polarization is reversed. This is the reason we see a strong SPP feature in ($\phi_2=25^\circ$, S light), which comes from diffraction direction $\phi = 85^\circ$. For diffraction directions between $0^\circ$ and $90^\circ$, the diffracted light has a component in both P and S light. Based on this discussion, we confirm the good agreement of the intensity in S and P polarized light from each mode is well explained.

There are also some discrepancies between the measurement and simulation:

Firstly, the measured TE waveguided mode is significantly weaker than indicated by the Setfos simulation. This is because Setfos uses optical constants measured on commercial ITO film, and the ITO in our experiment is from sputtering. Sputtered ITO has higher refractive index and larger absorption. TE waveguided mode is located
inside ITO layer. The absorption loss within ITO causes TE waveguided mode to damp away during propagation, and the actual TE waveguided intensity is weaker than the simulation anticipates.

Secondly, we assume in simulation when two diffracted modes intercept, their intensities add together. This is the case in most of the measured results with one exception. In ($\phi_2=25^\circ$, S light) around 2.3 eV and 3 $\mu$m$^{-1}$, TM waveguided mode and SPP mode intercept, and formed an anti-crossing feature. This phenomenon possibly indicates the two modes have a certain phase relation which results in destructive interference. Another explanation could be that the two modes split in a similar way bonding and anti-bonding hydrogen levels are formed. The exact reason of the phenomenon is still under investigation.

The author thanks Cheng Peng for his excellent work in Setfos simulation and Matlab coding, which explains the measurement result very well with optical theory.

**Quantify Light Extraction Amount**

With the information gained from ARES measurement, we can go back and look at the performance of a corrugated device. Based on the simulation, 30% of the light can escape to air mode. Consider a few factors that could reduce the device EQE, including quantum yield, charge balance, difference in ITO optical constants and slight deviation of layer thickness during evaporation, the 27% EQE is in good agreement with the prediction. Therefore, we can assume there are 14.4% light trapped in waveguide modes, 23.4% light trapped in SPP modes. By applying corrugation, the device EQE is improved to 33% (corresponding to a 22% enhancement). The additional 6% EQE comes from extraction of waveguided modes and SPP mode. Assume corrugation has
the same extraction efficiency for waveguided modes and SPP mode, this enhancement indicates about 16% of the trapped light is extracted from the device.

The enhancement in forward light intensity causes the current efficiency enhancement to be improved by 32%. This enhancement is bigger than the EQE enhancement amount (22%). There are two reasons for this difference: (1) The waveguided modes and SPP mode from -k and +k intercept at the center of air cone, causing the local extraction intensity at θ = 0° to be higher than the averaged extracted amount. (2) Current efficiency is dependent on the human eye response to different colors, thus is dependent on the emission spectrum. The corrugation enhances at 540 nm through TM waveguided extraction. This wavelength matches with the peak wavelength human eye has the strongest response to (555 nm). The TE waveguided and SPP mode enhancement contribute less to the forward current efficiency because the enhancement wavelengths (590 nm and 630 nm) are off the human eye response peak.

We compare the forward EL spectrum of a corrugated device and a planar device, and plot out the enhancement amount by wavelength in Figure 5-17. The three peaks in enhancement ratio correspond to TM waveguided mode, TE waveguided mode and SPP mode. The SPP enhancement is the strongest (2.2 times) amongst the three. This can be explained from two aspects. First, more light is trapped in the SPP mode than waveguided modes, the extraction of SPP results in higher enhancement than waveguided modes. Second, the SPP mode is spatially localized on the surface of the metal electrode. Corrugation on the metal is very effective in extracting trapped SPP modes. Based on this finding, we can predict the 400 nm pitch corrugation will work the
best with a red emitter peak at 630 nm (when the same OLED structure is used). We can also calculate the required pitch for SPP to enhance at 520 nm in the normal direction. To improve the current efficiency at green region (520 nm), a 200 nm pitch corrugation is needed.

**ARES Study of Poly Crystal Device**

We also fabricated OLED devices on corrugated poly photonic crystal substrates (abbreviated as poly crystal device). For comparison, the poly crystal pattern has the same pitch of 346 nm (unit-to-unit distance of 400 nm) as the single crystal pattern. The corrugation depth is measured to be 60 nm. The OLED structure used in this comparison is similar to the one used to study single crystal device, with one difference being the Alq$_3$ is not doped with 10% Cs$_2$CO$_3$. This difference is purely due to legacy. The poly crystal device was studied before single crystal device, and the device structure wasn’t optimized at that time. The lack of Cs$_2$CO$_3$ doping only affects the electron injection, which causes the charge balance to be worse than the doped case. The difference has no effect on the optical structure of the device. The planar device used for comparison also uses the same structure without Cs$_2$CO$_3$ doping, making a paralleled comparison with the poly crystal device.

Figure 5-18 shows a picture of an operating pixel of the corrugated device. The pixel is uniform with some amount of haze on the edge. Different from the single crystal device where we can see noticeable glares, there is no directional glares for single crystal device. This is because although the single crystal pattern has a hexagonal symmetrical lattice within each grain, the overall diffraction effect of all the randomly oriented grains has no preferred direction. The performance of the device is shown in Figure 5-19. The corrugated device has the same turn-on voltage as the planar device.
at 2.5 V. The leakage current below turn-on voltage is around 200 nA/cm², similar for the planar and corrugated device. The planar device shows a peak current efficiency of 86 cd/A, and a peak EQE of 25%. The corrugated device shows a peak current efficiency of 105 cd/A, and a peak EQE of 28%. The enhancement in current efficiency and EQE for the corrugated device is 23% and 12% respectively. The EL spectra of the corrugated device shows higher intensity than the planar device at around 600 nm. We also look at the angular EL spectrum of the poly crystal device Figure 5-20. Different from the single crystal device, the poly crystal device has very small distortion in the angular EL spectra.

We conduct ARES measurement for both the planar and poly crystal device and show the result in Figure 5-21. The air mode of the planar device is homogeneous. The poly crystal device shows a broad enhancement from 2 μm⁻¹ to 10 μm⁻¹ at the peak emission wavelength (λ = 520 nm). This is difference between the single crystal device, for which the air mode has multiple diffraction features due to waveguided and SPP mode extraction. To understand the origin of the diffraction feature seen in poly crystal device, we plot out its normalized air mode in Figure 5-22. Only one set of diffraction features are observed which intercept at the air cone center at 2.0 eV. This confirms the diffraction is from SPP mode extraction. However, this diffraction feature is singular, unlike the 3 branches seen in single crystal device. The reason has been discussed above: the overall diffraction effect of all the randomly oriented grains has no preferred direction. In a way, this feature is the integration of the diffraction features in single crystal device with φ from 0 to 2π. Therefore, the feature is also wider in k axis than in the single crystal case. The majority of energy is located in the ‘front’ of the feature, due
to diffracted light from small $\phi$ angles, while a weaker tail comes from the integration of diffracted light from large $\phi$ angles.

In the air mode, we are missing the waveguided mode diffraction features. This is because the mold used to make this corrugated substrate is shallow (60 nm), so the extracted waveguided mode intensity is relatively weak. Only SPP mode can be observed because it is stronger than the waveguided mode. To demonstrate the poly crystal substrate is also effective on the waveguided modes, we later switched to a new mold with depth of 120 nm and a pitch of 250 nm. After planarization, the corrugation depth is close to 80 nm, which ensures the device to have low leakage current. Because the pitch of the new mold is small, its diffraction vector $k_G$ is quite large (25.1 $\mu$m$^{-1}$). This causes the diffracted waveguided and SPP modes to intercept at $E_0 > 2.6$ eV ($\lambda < 480$ nm) in the center of air mode (Figure 5-23). We can clearly see three set of distinguished diffraction features coming from SPP mode, TE waveguided mode and TM waveguided mode. This confirms the poly crystal structure can extract waveguided modes as the single crystal substrate.

Although the corrugation depth is larger for the new mold, and the diffraction features from all trapped modes are observed in the air mode, its efficiency is actually lower than the planar device. One probable reason is that the photons emitted by the green emitter has $\lambda > 500$ nm, two times longer than the corrugation pitch, therefore the diffraction efficiency of the corrugation pattern is relatively weak. In addition, the corrugated device has some of leakage current, which could lower the baseline IQE and cause the overall EQE to be lower. The interaction between corrugation pitch and
diffraction efficiency is one important aspect of the research. It will be studied in the future work to find the optimal pitch.

**Discussion of Single and Poly Crystal Device**

In display or lighting application, light extraction techniques are important because they enhance the device efficiency, reduce power consumption, and improve device lifetime (by driving at a lower current to obtain the same brightness). Single and poly crystal corrugation can both improve the device efficiency, but there is a very important difference between the two methods. Single crystal OLEDs have strong angle dependent spectrum. When used for display purpose, their CIE coordinates will shift at different viewing angle; when used for lighting purpose, the directional glares also undermine the quality of the light. These properties make single crystals unsuitable for either application.

Poly crystal OLED has the important advantage of no glare and very small color shift at different viewing angles. Upon further optimization of pitch and depth, the efficiency enhancement amount of a poly crystal OLED can be match that of a single crystal device. There is big potential for poly crystal to be used in multiple OLED applications.

The author wants to thank Monica Samal for her excellent work in planarizing deep single crystal corrugation and fabricating stable, uniform OLED devices, and thank Dong-Hun Shin for his high quality 250 nm pitch poly-crystal corrugation mold.
Figure 5-1. Fabrication of corrugated OLED from a master mold through nano-imprinting.

Figure 5-2. OLED structure of the corrugated device fabricated on a single crystal substrate and a photo of the pixel driven at 1.25 mA/cm²
Figure 5-3. (a) J-V curve, (b) current efficiency plot, (c) EQE plot and (d) forward EL spectrum of a corrugated OLED device fabricated on a single photonic crystal substrate. The planar device is used as a reference.
Figure 5-4. Angle dependent EL spectra of a corrugated device fabricated on a single photonic crystal substrate.

Figure 5-5. Air mode of a corrugated device fabricated on a single photonic crystal substrate. It has a similar background as the planar device air mode, with additional features. The device is measured at a random substrate orientation $\phi$. This angle is later calibrated to be 5°.
Figure 5-6. Processing the air mode by dividing emitter spectrum to achieve the normalized air mode.

Figure 5-7. Normalized air mode for a single crystal device which reveals diffraction features from trapped modes.
Figure 5-8. 3D drawing of an OLED device and its corresponding mode distribution in $k_x$-$k_y$-$E$ space.
Figure 5-9. The diffraction of SPP mode by a 1D grating along the $k_x$ axis.

Figure 5-10. Top down and side view of the 3D diffraction along the $k_x$ axis seen by the $k_x$-$E$ plane.
Figure 5-11. The diffraction of SPP mode by a 1D grating at an arbitrary angle in the $k_x$-$k_y$ plane.

Figure 5-12. Top down and side view of the 3D diffraction in the $k_x$-$k_y$ plane seen by the $k_x$-$E$ plane.
Figure 5-13. Polarized normalized air mode measured at $\phi_1 = 5^\circ$. 
Figure 5-14. Polarized normalized air mode measured at $\phi_2 = 25^\circ$. 
Figure 5-15. Polarized normalized air mode simulated for $\phi_1 = 5^\circ$. 

$\Phi_1 = 5^\circ$

**S Light**

**P Light**
Figure 5-16. Polarized normalized air mode simulated for $\phi_2 = 25^\circ$. 

$\Phi_2 = 25^\circ$
Figure 5-17. Forward EL spectrum of a single crystal device and the enhancement ratio correlated to each extracted mode.

Figure 5-18. OLED structure of the corrugated device fabricated on a poly crystal substrate and a photo of the pixel driven at 1.25 mA/cm².
Figure 5-19. (a) J-V curve, (b) current efficiency plot, (c) EQE plot and (d) forward EL spectrum of a corrugated OLED device fabricated on a poly photonic crystal substrate. The planar device is used as a reference.
Figure 5-20. Angle dependent EL spectra of a corrugated device fabricated on a poly photonic crystal substrate.

Figure 5-21. Air mode comparison of (a) a planar device and (b) a poly crystal device.
Figure 5-22. Normalized air mode of a poly crystal OLED with $\Lambda = 340$ nm and $d = 60$ nm.

Figure 5-23. Normalized air mode of a poly crystal OLED with $\Lambda = 250$ nm and $d = 80$ nm.
We have used ARES study to investigate the mode extraction in single and polycrystal devices, and achieved current efficiency enhancement of 32% and EQE enhancement of 22%. The enhancement amount is not still satisfactory considering the added cost to make a corrugated OLED. In this chapter, we discuss the factors that affect the diffraction efficiency of corrugation.

**Corrugation Depth**

The diffraction efficiency of a grating is dependent on the corrugation depth. Larger depth is beneficial to achieve higher diffraction efficiency, but also introduces unnecessary complexity in electrical properties. The organic layer thickness of an OLED device is about 120 nm. When deposited on a corrugated substrate, the organic layers become corrugated as well. Deep corrugation causes thin area between the two electrodes, increasing leakage current level, and sometimes results in shorting problems. When the corrugation is too deep, it also introduces shadow effect, causing poor coverage of the sputtered or evaporated layer. We have observed when sputtering 70 nm ITO as the anode on a deep corrugation, the conductivity of the ITO strip is very low, due to discontinuity in ITO layer.

In our research, the photonic crystal molds generally have a depth larger than 120 nm. These molds have strong diffraction effects seen by naked eyes, but cannot make devices with good uniformity and stability. Therefore, a planarization step is done before ITO sputtering. Planarization is carried out by spin coating PEDOT onto corrugated glass substrate prior to ITO sputtering. PEDOT has a refractive index close...
to the glass substrate (n = 1.5), which doesn’t change the refractive contrast in the ITO/glass interface. First, the corrugated substrates go through a 5 min UV treatment to reduce surface energy, then PEDOT is spin-coated onto the substrate at 500 rpm for 40 seconds. This coated substrate was annealed at 150 degrees for 1 hour to remove the solvent. For very deep corrugation profiles, multiple steps of planarization are needed to control the depth. The planarized substrate has reduced peak-to-valley height, and the OLED devices fabricated on planarized substrate show good uniformity and low leakage current.

We study the effect of corrugation depth by controlling spin coating layers. The single crystal substrate is planarized by 1, 2 and 3 steps for comparison. Each planarization step is done by spin-coating 1500 rpm and post-annealing. SEM images indicate the 1 step corrugation has a depth of 95 nm, 2 step corrugation has a depth of 80 nm, 3 step corrugation has a depth of 65 nm. The OLED device current efficiency is shown in Figure 6-1. All the devices including the control device use the same OLED structure. The devices are not optimized in charge balance, so the current efficiency is half of the highest efficiency device. Nonetheless, we can look into the optical effect of the corrugation by comparing the relative efficiency. The planar device shows 40 cd/A current efficiency, the 3-step planarized device shows 55 cd/A current efficiency, the 2-step planarized device shows 60 cd/A current efficiency, and the 1-step planarized device shows 75 cd/A current efficiency. All the pixels are uniform due to moderate corrugation depth. The trend of increasing current efficiency with decreasing the planarization steps is expected.
The ARES measurement result for the devices are shown in Figure 6-2. The planar device has a uniform and featureless air mode. The corrugated devices all show clear TM waveguided mode and SPP mode features. TE waveguided mode feature is relatively and too close to the SPP mode thus difficult to distinguish. The intensity of the diffraction features is noticeably stronger when the planarization step is reduced. This trend is also present in the forward EL spectrum Figure 6-3. The diffraction peaks height increases as the planarization step decreases, which agrees with the improved current efficiency we measured.

Based on these results, we are encouraged to further increase the corrugation depth for stronger diffraction. This is achieved by using 1-step planarization, and increasing the spin-coating speed of PEDOT to 2500 rpm and 4000 rpm. Planar device and the same 1500 rpm PEDOT 1-step planarized device are used as references. As a result, the planar and 1500 rpm devices both have uniform pixels and good device stability; the 2500 rpm device has non-uniform pixel area due to poor coverage of the planarization; the 4000 rpm device has the same issue of non-uniformity pixel, with additional high leakage current and very fast degradation. We compare the measured ARES results in Figure 6-4. Using SPP diffraction feature intensity as a criterion, we can see a monotonous increase in diffraction intensity. This confirms higher corrugation depth causes stronger diffraction, but could potentially harm the device stability and introduce leakage current. The balanced corrugation depth with our current single crystal mold is around 95 nm.
Index Contrast

The diffraction efficiency of a corrugation is also a function of the refractive index contrast (Δn) at the corrugated interface. Intuitively thinking, when Δn = 0, the corrugated boundary ceases to exist, and light will not experience any diffraction. Therefore, a larger Δn can cause strong diffraction. In our OLED devices, most of the diffraction happens at reflective metal/organic layer interface and ITO/glass interface.

A schematic drawing of the planarization layer is shown in Figure 6-5. When PEDOT is used as the planarization layer, the index contrast at the ITO/glass interface is unchanged. Although the index contrast is great at the metal/organic interface (organic materials have indices of 1.75, and metal has very small refractive index in visible wavelengths), it cannot affect optical modes of which the electric field is not near the metal surface. In the OLED device we study, SPP mode is on the surface of metal, TM waveguided mode is distributed in the organic layers, TE waveguided mode is distributed in the ITO layer. The corrugated electrode can affect the SPP and TM waveguided mode, but is powerless against TE waveguided mode because most of its energy is in the ITO layer. TE waveguided mode is mostly diffracted by the interface between ITO and glass, which has an index contrast of 0.5 at λ = 520 nm. This is the main reason we see very little amount of TE waveguided modes features with our corrugated substrates.

To effectively extract TE waveguided mode, we need to increase the index difference at the ITO interface. We obtained a solution processed low index dielectric material from SBA Materials which has a refractive index of 1.2. The low refractive index is controlled with film annealing temperature (125°C ~ 450°C) and time (30 mins ~ 2
hours). The low index layer is used as the planarization for comparison with PEDOT. The index contrast at the glass/ITO interface is increased to 0.8 with low index layer.

We fabricated OLED devices on low index material planarized corrugated substrates (low index devices), and compare it with OLED fabricated on PEDOT planarized corrugated substrates (PEDOT devices). The PEDOT planarized device (Figure 6-6) shows strong extraction of TM waveguided mode and SPP mode, due to the strong diffraction by the top electrode. There is TE waveguided mode feature, but the intensity is rather weak compared to the other two modes. The low index device (Figure 6-7) on the other hand shows very strong TE waveguided mode diffraction feature, while the TM waveguided mode and SPP mode diffraction is weak.

The difference in the extraction efficiency by PEDOT and low index planarization can be explained as follows: PEDOT only requires 20 min of thermal annealing, after spin-coated on the corrugated epoxy, it quickly solidifies and follows the wavy surface with small amount of planarization effect; the low index material requires over 1 hour annealing, after spin-coated to the corrugated surface, it has enough time to relax to the valleys of the corrugated pattern and reduces the corrugation depth. As a result, the corrugation depth on the top electrode is larger for PEDOT planarization than low index planarization, causing the diffraction of TM waveguided mode and SPP mode to be strong. Although the low index planarization reduced the corrugation depth, the effect is overcome by the large index contrast between ITO and low index layer, causing strong diffraction of the TE waveguided mode.
Low index layer between ITO and glass interface is proven to be very effective to extract TE waveguided mode. In the future work, we will optimize the processing method of low index material, and maintain the large corrugation depth for TM waveguided mode and SPP mode extraction. We expect to see the EQE of a corrugated device to be improved to 36% with fully optimized corrugation with low index layer.
Figure 6-1. Current efficiency comparison of a planar device and corrugated devices with 1, 2 and 3 steps of planarization
Figure 6-2. Normalized air mode comparison of a planar device and corrugated devices with 1, 2 and 3 steps of planarization

Figure 6-3. Angular EL spectra comparison of a planar device and corrugated devices with 1, 2 and 3 steps of planarization
Figure 6-4. Normalized air mode comparison of a planar device and corrugated devices with 1500 rpm, 2500 rpm and 4000 rpm 1-step planarization.

Figure 6-5. Schematic drawing of corrugated substrate (a) without planarization (b) with PEDOT planarization (c) with low index material planarization.
Figure 6-6. Normalized air mode of a corrugated device on PEDOT planarization layer measured at two device orientations.
Figure 6-7. Normalized air mode of a corrugated device on low index planarization layer measured at two device orientations.
This research is entirely dedicated to the light extraction of OLED with corrugation. By the combined power of OLED device fabrication, ARES optical analysis and Setfos simulation, the true optics behind light extraction is revealed by the study, much like how trapped light is extracted from the OLED device.

Corrugation using periodic photonic crystal pattern diffracts trapped light by reducing its in-plane wavevector across the air cone. TM waveguided mode, TE waveguided mode and SPP mode features have been observed in both single and poly crystal devices. For the single crystal device, we were able to demonstrate a 22% enhancement in EQE from 27% to 33%, and 32% enhancement in current efficiency from 91 cd/A to 120 cd/A. The extraction efficiency is estimated to be 16%. For the poly crystal device, a 12% enhancement in EQE and a 23% enhancement in current efficiency is observed. The poly crystal device shows very small angular EL distortion and no glares, suitable for various display and lighting purpose.

To achieve to fullest power of extraction from a corrugated substrate, in-depth analysis is conducted on the factors that could affect the extraction efficiency. Corrugation depth plays a key role determining the EQE of a corrugated OLED. Sufficient depth is needed to diffract trapped lights, while too much depth is detrimental to the device electrical stability. A fine line of 100 nm depth is discovered which results in high diffraction efficiency and relatively low leakage current.

Index contrast is essential in extracting waveguided mode, especially TE waveguided mode which is located inside ITO layer. By replacing PEDOT with low
index material, the index contrast at the ITO/glass interface is increased from 0.5 to 0.8. As a result, the mysterious TE waveguided mode diffraction becomes significant.

The future work of this research is listed as follows:

First, the EQE of the corrugated device needs further optimization. Low index material with refractive index close to air \((n = 1)\) will bring in extra power to extract waveguided modes. To take full advantage of the index contrast, more light needs to be funneled into the TE waveguided mode. This can be done with high index HTL and HIL material, which works as optical mode sink to attract photons. Corrugation profile hasn’t been fully studied in this research, which can also significantly affect diffraction efficiency. Sinusoidal shaped or dome shaped corrugation unit is not optimal for high diffraction efficiency. In the future work, we will explore triangle shaped and parabola shaped corrugation. A fully optimized corrugated OLED is expected to have over 50% diffraction efficiency and over 45% EQE. When a half-ball lens is attached to extract substrate mode, the EQE should be increased to 80%.

Second, we will study SPP emission OLED which has ultra-high forward luminance. From our result, the current efficiency of the corrugated devices always shows higher amount of enhancement relative to the total EQE improvement. This encourages us to design OLED devices which has strongest light output in the forward direction. This is particularly useful for retina display techniques where the display is close to the human eye, and the image is directly projected to the retina at a fixed direction (e.g. virtual reality headset display). We have observed very strong enhancement from SPP diffraction. For the next step we will shift the SPP peak to the emission peak of emitter, and funnel all the photon energy to SPP mode. This can be
done by replacing the top electrode with high conductivity, low absorption loss metal (e.g. silver). The emitter also needs to be located as close as the electrode as possible. High current efficiency as much as 200 cd/A using Ir(ppy)$_2$(acac) is expected using SPP emission.

There are other important aspects of corrugated OLED left untouched in research, including device lifetime study, color coordinate shift, and its performance on flexible substrate. Each of these aspects will be included in the future work, making it a complete study of corrugation.
LIST OF REFERENCES


BIOGRAPHICAL SKETCH

Xiangyu Fu (付相宇) was born in Dalian, Liaoning, China. He graduated from Fudan University, Shanghai with a B. S. in optics in 2011. In the same year, he was admitted to the Material Science and Engineering Department at the University of Florida. In 2012, he joined Dr. Franky So’s group to pursue a Ph. D. degree, and focused his research on light extraction techniques in OLED devices.